Black Carbon Deposition onto the Greenland Ice Sheet: Particle Quantification and Characterization

Senior Thesis

Submitted in partial fulfillment of the requirements for the

Bachelor of Science Degree

At The Ohio State University

By

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August 2015

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Abstract

Black carbon (BC), resulting from the incomplete combustion of biofuels and fossil fuels, has become the second most influential climate forcing mechanism on Earth (Bond and Sun, 2005). As variations in BC morphology reflect emission source, the examination and quantification of BC in glacier ice and snow may reveal spatial and temporal trends of the origin and abundance of BC in arctic ecosystems. This study analyzed BC particle size distribution, concentration, and morphology of twelve samples from Leverett Glacier, Greenland, in order to evaluate the magnitude and influence of specific anthropogenic activities on the carbon cycle. The results indicate four substantial findings that (1) the deposition of BC particles larger than 0.3μm is greater in fresh snow than in older ice, (2) the majority of BC particles measuring >0.3μm are smaller than 1μm, (3) BC particle size and abundance exhibit an inverse relationship, and (4) BC emissions from biofuel burning dominates the 2012 snowpack. These findings are important as they indicate that the incomplete burning of biofuels including coal and wood has significant, detrimental influences on climate and public health. However, the reduction of BC emissions globally by, for example, efficient biomass and fossil fuel burning methods, would decrease these deleterious BC-mediated effects.

Acknowledgements

I extend gratitude to my research advisor, Dr. Joel Barker, whose guidance and assistance made this project possible. I hold in the highest regards his ambition, consistent guidance, words of assurance, and willingness for collaboration. Along with Dr. Barker, appropriate recognition for Dr. Jemma Wadham of the University of Bristol, is essential for providing the Leverett Glacier samples that formed the basis of this study. Further appreciation is given to The Ohio State University, which enabled this research through the former Climate, Water, and Carbon Seed Grant Program. Many acknowledgements also go out to the number of exceptional School of Earth Sciences professors that I have had the pleasure of learning from during the course of my undergraduate education. Due to their knowledgeability and enthusiasm, I was able to persevere through any obstacles and remain passionate in my studies. And finally, a special and loving thank you to my parents, whom are responsible for not only my education and ability to attend The Ohio State University, but also my entire livelihood. I cannot fully express how their encouragement and support, along with other family and friends, has positively influenced my life and successes.

Figures/Images

Tables/Charts

Introduction

Atmospheric aerosols resulting from industrial and agricultural expansion have increased since the Industrial Revolution approximately 175 years ago (Watson et al., 1990). Some of these aerosols, for example black carbon (BC) particles or sulphate aerosols, are radiatively active and may affect global climate (Watson et al., 1990). The majority of studies conducted thus far have focused on atmospheric CO_2 as a primary factor influencing global climate change (Bond and Sun, 2005). However, an examination of other atmospheric aerosols, may provide a more holistic view of Earth's sensitivity to society's expansion, urbanization, and industrialization.

BC is a key factor influencing global warming and is second only to increasing atmospheric CO_2 concentration in contributing to global climate warming since the Industrial Revolution (Bond and Sun, 2005). The global temperature response to BC is as much as three times greater than that of equal CO_2 forcing (Flanner et al., 2007). Resulting from the incomplete combustion of fossil fuels and biomass burning, BC emitted into the atmosphere is a global pollutant and facilitates warming of the Earth by absorbing incoming shortwave radiation within the troposphere (Chang, 2011). BC is also the most absorptive impurity per unit mass in snow and indirectly contributes to global warming (Doherty et al., 2010). Acting as a positive climate-forcing feedback loop, the addition of BC impurities in arctic environments result in albedo reduction and increased radiation absorption. In turn, the troposphere warms and facilitates sea ice/ glacier melt. Further, as ice and snow melt, ice and snow grains coarsen resulting in a larger effective radius that exacerbates the absorption of shortwave radiation (Flanner et al., 2007). Although BC is effective as a climate forcer, the formation of BC from incomplete combustion, instead of direct energy use as with CO_2 and other aerosols, enables

more obtainable and effective mitigation strategies (Chang, 2011). As a result, emissions can be reduced by development of more efficient biomass/fossil fuel burning methods and does not require limiting of the specific anthropogenic activities themselves.

Prior to deposition, atmospheric BC may travel for weeks and over thousands of kilometers, in the atmosphere before removal by precipitation (Clarke et al., 2004). The ability of BC to travel significant distances from its emission source suggests that arctic regions may accumulate evidence of biomass burning and fossil fuel combustion from as far as North America, Europe, and Asia (McConnell et al., 2007). The progressive accumulation of snow and ice during glacier formation enables glaciers to function as valuable climate proxies by providing a comprehensive archive of atmospheric particulates that are deposited onto their surfaces over time. More precisely, precipitation deposited onto the glacier surface with each succeeding winter season will represent a younger relative age to the underlying layers.

Ice taken from alpine glaciers or the Greenland Ice Sheet (GIS) can preserve chemical and physical characteristics that serve as a record and can be examined to deduce past atmospheric composition (Chylek et al., 1995). The measured abundance and concentration of pollutants deposited in remote arctic environments, far from pollution sources, offer valuable insights on anthropogenically-derived particulates that may be distributed over long distances and thus may represent trends that can be extrapolated over the northern hemisphere. Deposits of BC onto glacier ice can be examined to determine both the relative abundance of BC deposited from the atmosphere and potential BC source by examining particle morphology (Doherty et al., 2010).

Background

BC is the byproduct of the incomplete combustion of organic material, and the quantity of BC is contingent upon the efficiency and amount of biofuel or fossil fuel burned (Novakov et al., 2003). Incomplete burning of biomass such as wood and coal or during forest, residential, and agricultural fires, accounts for approximately 65% of all BC emissions globally (Chang, 2011). The remaining BC, resulting from combustion of fossil fuels for transportation, industry, heating, etc (Doherty et al., 2010). Kasparian and others (1998) in a study of BC particle morphology found that particles vary with carbon source and combustion intensity. For example, char, produced during biomass burning, will characteristically have an irregular shape and rough surface texture while, soot, has spherical and smooth features and is formed during fossil fuel combustion. Atmospheric BC residence time ranges from 3.7 to 23.2 days (Jaio et al., 2014) and varies as a result of atmospheric moisture composition; snowfall is considered to be one of the most efficient removal processes (Hadley et al., 2010). BC particles from North America may travel significant distances from their emissions source before being scavenged by precipitation. Backward air mass trajectories of aerosols in Greenland, have shown that emission source regions differ by season and suggests that BC emissions from North America dominate in the fall and are also significant in the winter (Davidson et al., 1993).

Goals and Objectives

Over the past 100 years, global temperatures have risen between 0.6°C and 0.9°C. Even more shocking, the rate of global warming during the latter half of the 20th century is double that of the first half (Riebeek, 2007). As the Earth continues to warm at an unprecedented rate (Watson et al., 1990) the need to achieve a better understanding of the factors contributing to global warming and to identify appropriate and effective mitigation strategies is clear. In order to accomplish this, a comprehensive assessment of BC sources and magnitude of particle deposition is required. This study utilizes optical analytical methods to quantify BC deposition from the atmosphere onto GIS and to determine the source BC emission based on the specific morphological features from ice and snow of different ages from Leverett Glacier, Greenland.

Field Site

Leverett Glacier (67° 06′N, 50° 09′W) is an outlet glacier flowing from Southwestern GIS (Figure 1). The GIS, is the second largest polar ice cap on Earth, spanning 1,710,000km^2 and reaches an altitude of 3,500m at its highest point (NSIDC, 2015). Due to the systematic accumulation of precipitation on Greenland, the ice sheet functions as an invaluable record for northern hemisphere climate over the past ~100ka (Rasmussen et al., 2013), and is perhaps the only remaining Pleistocene glaciation in the Northern hemisphere.

Approximately 85km in length, Leverett Glacier is a result of snow accumulation, firnification, ablation, and deformation. Leverett Glacier is located on the periphery of the GIS which is associated with a higher number of melt days compared to the interior of the ice sheet, has downward sloping topography, and resides on top of smooth, Precambrian crystalline rock. These characteristics indicate climactic change vulnerability as melt seasons begins earlier and are more severe with global warming, and also facilitates the relatively rapid west-southwest flow of ice and meltwater across the 600km^2 drainage basin from the high elevation of the interior, to the sea (Tedesco et al., 2015).

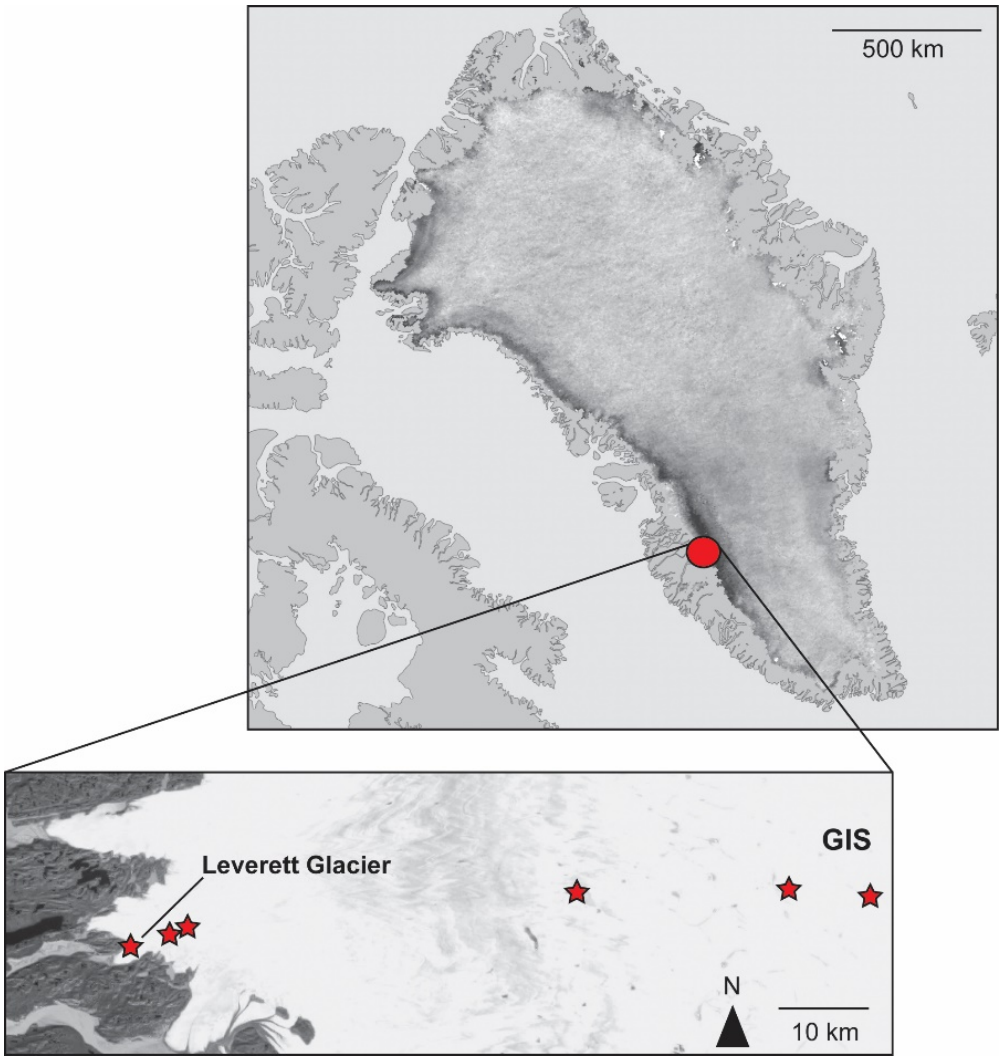

Figure 1*. Leverett Glacier, Greenland and sampling locations. Samples from left to right: 10, 3, 7, 8 (A, B, & C), 9. The final star marking location of samples 1, 2, 4, 5, and 6 (Google Earth, 2015).*

Sample Description

Twelve ice and snow samples analyzed in this study were collected along a 70km transect extending from the Leverett Glacier terminus to above the equilibrium line and onto the GIS during the summer of 2012. The samples preserve a record of atmospheric BC from North America via the North Atlantic Oscillation flow of the atmosphere (Christoudias et al., 2012). Glacier ice formation and flow processes result in samples that are collected closer to the terminus being older than those taken at higher elevations (Benn and Evans, 2010). Surface snow collected above the equilibrium line record BC deposited during the preceding winter (2011–2012). The nature of glaciers to accumulate and move in this way enables Leverett Glacier to function as a long-term temporal archive of BC that has traveled weeks from North America before deposition. Figure 1 shows the location of Leverett Glacier within the GIS and the sampling locations. The sample farthest west, at the terminus of the glacier, sample 9, is the oldest of all samples and is of snow taken 35cm from the surface. In sequence towards the east, next is sample 3 (3km from the glacier terminus), sample 7 (7km from terminus), sample 8 (8km from terminus; 8A: snow, 8B: deeper snow, 8C: ice), and sample 10 (50km from terminus, surface snow). The remaining star marks 70km from the terminus and includes the youngest samples 1, 2, 4, 5, and 6.

Methods

Standard Collection

As a preliminary step to quantify Leverett Glacier BC, two standards of reference, both representing distinct BC emission sources were produced and collected. BC produced by hydrocarbon combustion from a 2013 Lexus RX 450h gas powered automobile was collected by passing engine exhaust through a glass fiber filter paper (GF/F; 0.7μm nominal pore size) under vacuum. BC produced by vegetation burning was collected by passing smoke from burning leaf litter gathered from outside of Bryd Polar Climate and Research Center through a glass fiber filter paper under vacuum. The BC retained on each filter paper was examined under a Nikon Eclipse 80*i* H550L stereo-microscope to determine BC particle size and morphology. The establishment of known examples of char and soot, references A and B respectively, was a tactic to diminish the uncertainty in distinguishing BC from other debris during analytical methods of the glacial ice. Images 2 and 3 provide examples of the two distinctive BC morphologies observed within this study.

***Image 2**. Char particle/reference A (approximately 1.5µm in length) obtained through vegetation burn. Note rough surface texture.*

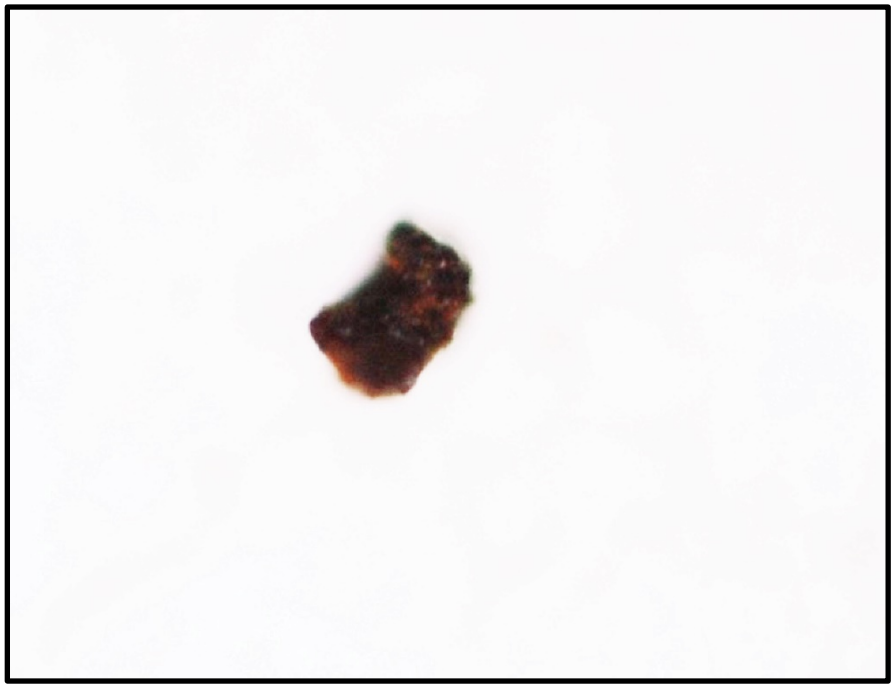

***Image 3.** Soot particle (approximately 0.75µm in length) obtained from sample 9. Globular/smooth features in resemblance of reference B.*

Sample Analysis

The twelve samples of ice and snow from Leverett Glacier were kept frozen at -23°C until analysis. A known mass of ice or snow was weighed using an Explorer Pro EP613C analytical balance, placed into a sterile jar, and left at room temperature in the dark until fully melted. All samples where then filtered under vacuum through a glass-fiber membrane filter paper (GFMF; 0.3μm nominal pore size) using a clean glass filter tower. Wet filters were then dried and examined using a Nikon Eclipse 80*i* H550L stereo-microscope under 10X objective lens power.

Particles identified as BC were recorded on a basis of long axis length using a 0.150mm absolute-scale reticle, or 0.0015mm relative-scale reticle when used in conjunction with a 10X objective lens. Morphological features in resemblance to standard A or B were also documented for noteworthy particles. An assumption was made that particulate matter was evenly distributed across the GFMF paper for all samples. Therefore, half of the filter was examined and particle abundances associated with specific diameters were multiplied by two. Concentration was found by dividing the total number of BC particles per sample by the volume of the sample filtered. Utilizing this methodology resulted in BC particle abundance, size distributions, and total BC (soot and char) concentrations of the samples.

Results

Of the twelve ice and snow samples collected from Leverett Glacier, BC particle sizes ranged from 0.375µm to 22.5µm, by basis of long-axis. Nominal BC particles less than 0.3µm were not observed in this study due to the restrictive GFMF paper pore size. The largest particle, 22.5µm, was observed in sample 7 and had globular/smooth features that are distinctive of incomplete fossil fuel combustion and representative of standard B. An overall downward trend in particle abundance with increasing particle size for all twelve samples is presented in chart 1 and is indicative of an inverse relationship between size and abundance. The remaining charts (2 and 3) depict BC particle distribution for samples located at the same distance from glacier terminus which allow for easier comparison and also follow the same inverse relationship. Additionally, within the twelve samples it was observed that BC particles less than 1µm resembled standard of reference A (image 2), whereas the larger particles were more closely related to reference B and image 3.

Concentration of BC particles/ml of each Leverett Glacier sample are given in table 4. The majority of sample BC concentrations fall within 9.0 particles/ml and 13.0 particles/ml and includes samples 1, 2, 3, 7, 8A, 8B, and 8C. However, higher quantities were also measured within this study, and reach a maximum of 52.54 BC particles/ml for sample 10 which is located closest to the glacier terminus.

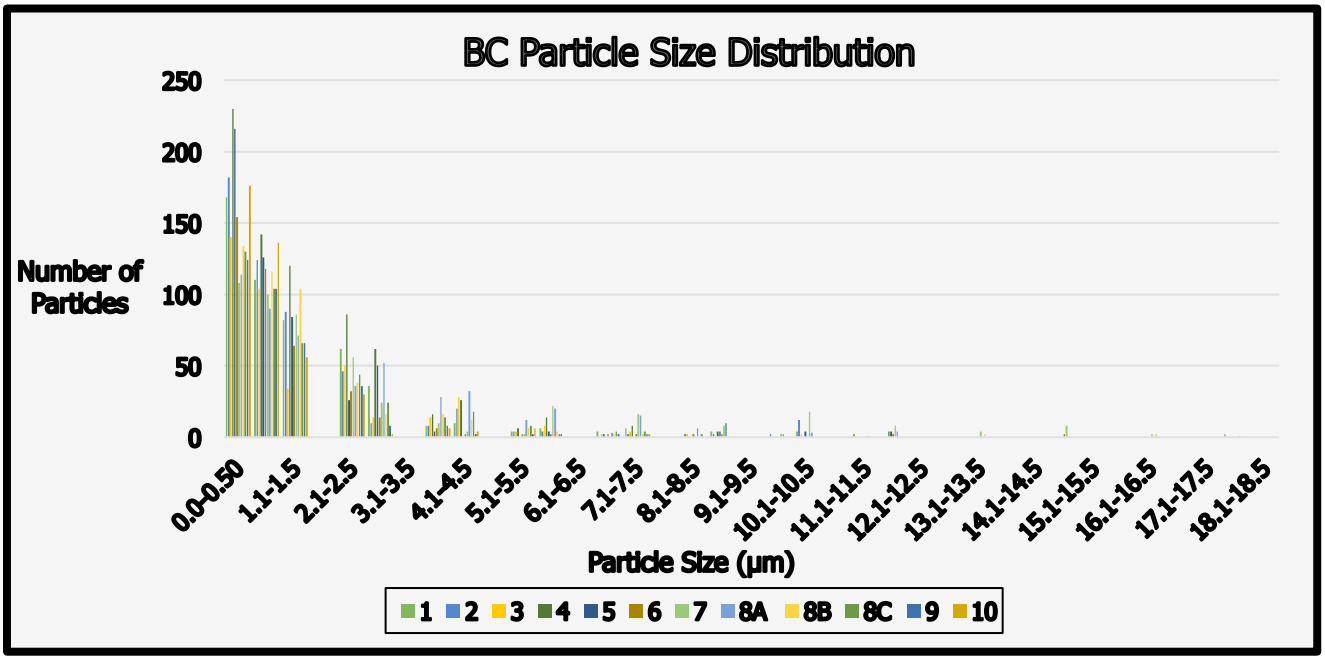

Chart 1. *Size distribution for all twelve samples across Leverett Glacier. Two additional particles for sample 7 at 19.1-19.5µm as well as at 21.1-22.5µm.*

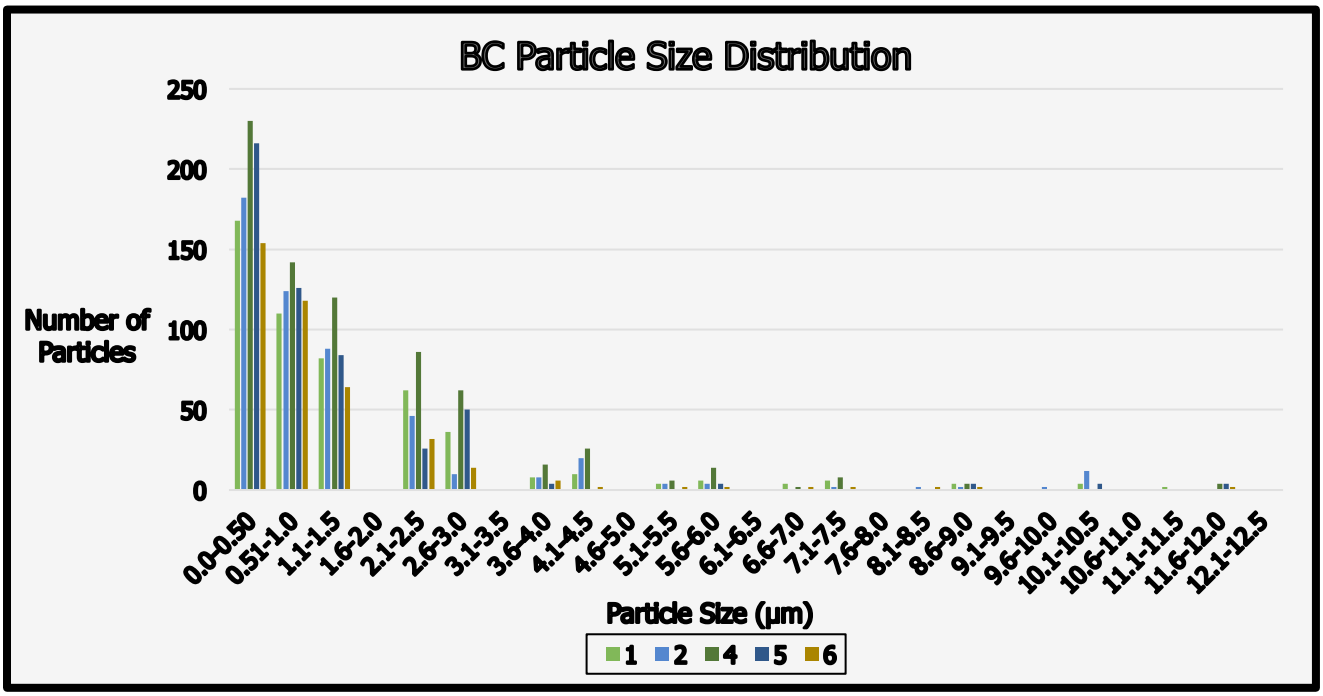

Chart 2*. Size distribution of samples taken at 70km from glacier terminus. Two additional particles for sample 1 at 17.6-18µm and two particles for sample 6 at 14.6-15µm.*

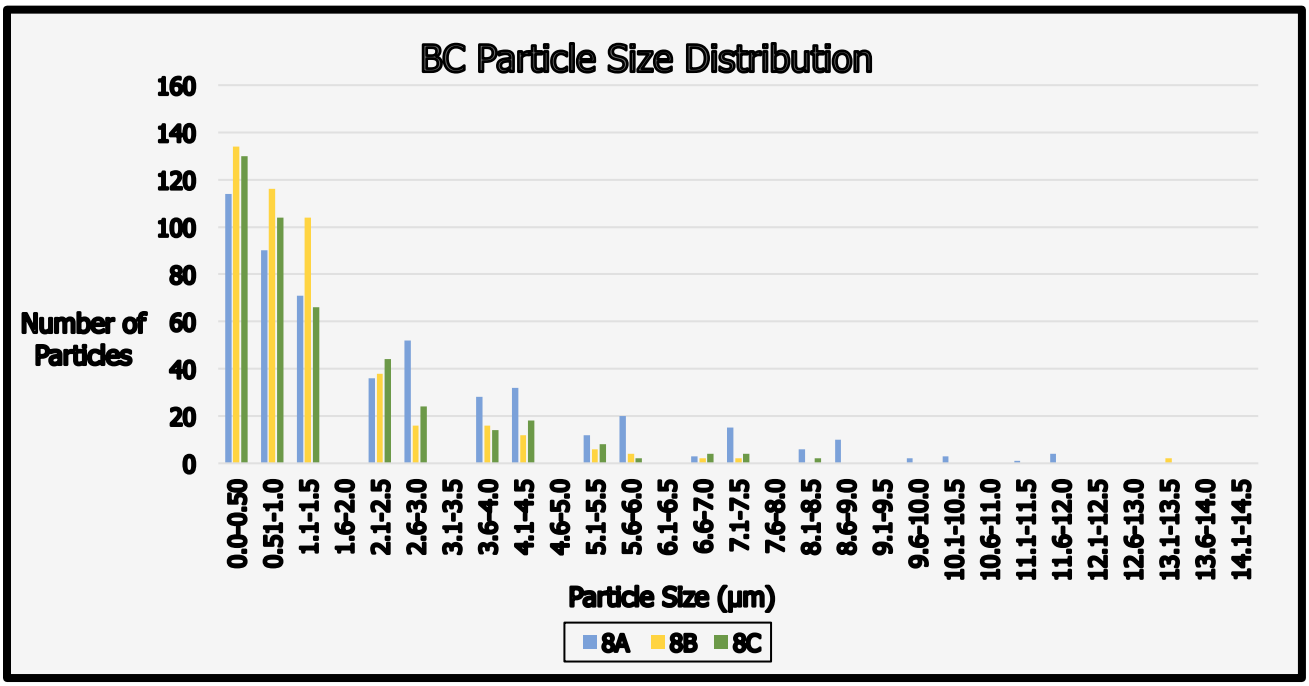

Chart 3. *Size distribution of samples taken 8km from glacier terminus. Additional particle for 8A at 17.6-18µm and two for 8B between 16.1-16.5µm.*

Table 4. *Summary of BC particles.*

Sample Number	BC Concentration (# of particles/ml)	Distance from Terminus (km)	Morphology Notes
1	12.34	70	
2	9.58	70	
3	11.91	3	large particle; B
4	29.81	70	abundant small particles; A
5	16.87	70	abundant small particles; A
6	19.17	70	
7	10.76	7	largest observed particle; B
8A	11.54	8	large particle; B
8B	11.58	8	
8C	11.04	8	
9	50.66	50	
10	52.54	terminal zone, 35cm into snowpack	

Discussion

Particle Abundance

A combination of firnification and glacier flow dynamics means that ice exposed at the glacier terminus is older than ice exposed near the equilibrium line (Benn and Evans, 2010) while snow exposed on the surface of the supraglacial snowpack during the summer season would have been deposited the preceding winter. BC emissions have increased as the result of global industrialization and landscape change associated with agricultural expansion (Chang, 2011). As a result, in general, one would anticipate higher BC concentration in post-industrial ice and snow than ice and snow formed prior to the Industrial Revolution. Of course, superimposed on this general trend will be more episodic variation in global BC emissions such as wars, economic upturn and recession, legislative restrictions, extensive wildfires, etc. While we were unable to assign ages to our ice samples, we would anticipate being able to observe a difference in abundance and/or BC source, as indicted by differences in BC particle morphology, between surface snow located above the equilibrium line (samples 1, 2, 4, 5, 6, 9) and ice located below the equilibrium line (samples 3, 7, 8A, 8B, 8C, 10).

Table 4 displays BC particle abundance for samples collected during this study. Overall, five out of six samples taken from below the equilibrium line depicted smaller BC concentrations than five out of the six samples taken from above the equilibrium line. This is supportive of the hypothesis that post-industrial ice BC concentrations will highlight growing anthropogenic emissions. Although samples 2 and 10 did not follow the general trend as the other eight samples, and there was no direct linear progression across the Leverett Glacier transect, it is possible that this is a reflection of irregular periods of BC emissions as explained in the previous paragraph.

Particle Size

BC particle size and abundance was the most evident result of this research. Out of the 5,698 BC particles observed throughout the combined twelve samples, approximately 57% were less than 1µm. In fact, in ten out of twelve samples, over 50% of total BC particles fell within the narrow 0.375µm to 0.75µm range whereas the remaining particles fell between the wider range 1.5µm up to 22.5µm. Comparable results are reported for urban aerosol studies which found peak aerosol abundances at 0.1µm and 0.9-1.0µm (Kasparian et al., 1998) or similarly, 0.09µm and 0.7-0.8µm (Fruhstorfer and Niessner, 1994). Interestingly, BC particles deposited onto the GIS will have been transported much further from their combustion source than urban BC aerosols are, suggesting that small BC particles can be transported long distances and their distribution does not depend on proximity to an emission source. There are many environmental and health implications associated with the finding that BC particles less than 1µm are transported globally. For example, small BC particles are more chemically active (Kasparian et al., 1998) and more efficient at reducing snow albedo while BC particles larger than 1µm have insignificant influence (Flanner et al., 2007). Further, smaller particulates are more significant to public health and are more detrimental to cardiovascular well-being and lung capacity (Kasparian et al., 1998; Chang, 2011).

Particle morphology

BC particle morphology can be used as a tool to determine BC source. BC particles with a smooth morphology are indicative of fossil fuel combustion (Image 3) while a rough texture is more indicative of a biomass burning source (Image 2). In this study, the largest particles were found at 3km, 7km, and 8km from the glacier terminus and had globular/smooth features that resembled standard B. Farther up the transect, at 70km, small particles characterized by rough surface texture with similarities to standard A were abundant. This suggests a transition from soot to char from terminus to equilibrium line, and is indicative that fossil fuel-source of BC predominates in older ice whereas BC resulting from biomass burning was dominant in the 2012 snowpack.

Sources of Error

As with all research, uncertainties are possible throughout the analysis process. Within this effort to quantify abundance and characterize morphology of BC deposits on Leverett Glacier, there are potential skepticisms to discuss. First and foremost, problems associated with sample preparation could lead to misinterpretations in data analysis. For example, BC is typically hydrophobic (Doherty et al., 2010) and so, could have remained in the sterile jar after ice melt or on the walls of the glass filter tower. This would ultimately result in inaccurate abundance and possible distribution results that were lower than the true value. Additionally, evaluation of only half of the glass-fiber membrane filter paper and assumption of even distribution of particulates across the entire filter for all twelve samples, it is plausible that there was some random error within the study. And finally, as the scanning electron microscope (SEM) was inoperable at the time of this study, determination of the elemental composition of particles was not possible and other particulates within the samples such as brown carbon and ash, could have been inaccurately accounted for as BC during purely optical analysis. If so, the provided BC abundance and concentration results may be an over-estimation despite representing accurate trends in the data.

Conclusions

The results of this study show that BC particles are deposited on Leverett Glacier and have generally increased in abundance over time. The majority of these particles are between 0.3µm and1µm and can be classified as char based on their morphology. This finding is important as it indicates that the incomplete combustion of biofuels including coal and wood is the most significant source of BC transported and deposited in the arctic and thus helps to target the highest BC-emitting activities. Equipped with the knowledge that coal and biomass burning are the dominant sources of BC, instead of petroleum burning, mitigation strategies can be implemented to reduce atmospheric BC. An example of effective mitigation strategies could include the development and distribution of more fuel-efficient stoves/heat sources and the implementation of land clearing techniques that do not involve biomass burning. In doing so, there is the potential to moderate BC emissions, and by extension global warming, by reducing the direct absorption of radiation by BC particles within the atmosphere and lessening the current ice-albedo feedback.

Suggestions for Future Work

In order to advance the assessment of the severity and ramifications of rapid industrialization, supplementary studies on BC emissions with the use of scanning electron microscopy with energy-dispersive X-ray (SEM/EDX) are necessary. Should analysis of the Leverett Glacier samples be done using a SEM/EDX approach, BC particles could be distinguished by elemental composition, eliminating some error associated with identifying individual BC particles based on morphology alone, and ultimately provide more accurate identification by supplementing the identification with chemical information. For example, the presence of high amounts of sulphur might be associated with fossil fuel or coal burning, while the absence of sulphur might suggest a biomass burning source. Preliminary analysis of our samples using SEM/EDX was initiated during this study, but instrument malfunction did not allow us to complete the analysis. Additionally, utilizing a GFMF paper with a reduced pore size, or applying a double filter method with different pore sizes as conducted in another arctic snow study (Doherty et al., 2010), would enable smaller, more radiatively active, BC particles around 0.1μm to be examined. Lastly, although snow/ice albedo is highly vulnerable to BC, research on other albedo- reducing impurities such as brown carbon, in conjunction with BC, may provide an enhanced representation of true arctic sensitivities.

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